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WATER DECONTAMINATION

Conrad P. Straub

Robert A. Taft Sanitary Engineering Center
Cincinnati, Ohio

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WATER DECONTAMINATION*

by

Conrad P. Straub**

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** Senior Sanitary Engineer, Robert A. Taft Sanitary Engineering Center, U. S. Public Health Service. Assigned to the Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Although radioactivity has been known since 1896 when Bequerel first observed the phenomenon by its effect on photographic plate, it has been of little concern or interest to the sanitary engineering profession until recent times.-- during the last decade in particular. Our interest and concern stems from the fact that large quantities of radioactive materials are produced during fission of certain naturally occurring heavy radioactive materials in nuclear reactors or piles. These radioactive fission products, many of high specific activity, being unstable, disintegrate releasing ionizing radiations which are damaging to living tissue, and as a result their release or discharge into the environment must be carefully controlled.

With the availability of a great variety of low cost radioisotopes many useful applications have been found in medicine, industry, and research. Naturally, this increased use has resulted in the discharge of these materials into the environment. A recently published survey of 1,027 users of radioisotopes(1) showed that about 41 per cent of them disposed of their

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radioactive wastes by dilution and discharge to the sewers. As additional peacetime applications are developed, increasing amounts of radioisotopes will probably be released. This increased discharge of radioactive materials is of concern to the sanitary engineer because he is responsible for maintaining a healthful environment.

At least one instance is known where death was attributed to the intake of a sufficiently large quantity of radioactive water. Gettler and Norris[2] reported a death due to pathologic lesions brought about by radium, which had entered the system through consumption of bottled drinking water over a period of several years. The water was said to contain two micrograms (μg) of radium in each 60 ml, and this is approximately equal to 0.033 microcuries per ml (μc/ml) or to a concentration approximately one million times higher than that recommended in NBS Handbook 52[3] for continuous intake of this material.

Maximum Permissible Concentrations (MPC)

NBS Handbook 52 is to the engineer engaged in radiation problems what the U. S. Public Health Service Drinking Water Standards are to the engineer concerned with certifying waters for use on interstate carriers. Both define recommended levels for specific contaminants, the former listing the MPC values for various radioisotopes in water, whereas the latter delineates chemical and bacteriological parameters. Anyone active in the field of radiological health should familiarize himself with the contents of NBS Handbook 52.

Not only does NBS Handbook 52 list the limiting concentrations in water and air for many of the radioisotopes of interest, but also states that in the case where the individual radioisotopes are not known, the permissible concentration of alpha, beta, and gamma activity in water shall not exceed 10^{-7} μc/ml. This value may be compared with emergency values recommended by the AEC for waters contaminated by radioactive materials following a nuclear bomb detonation which for ten days use immediately after the blast are 9×10^{-2} μc/ml for beta-gamma activity and 5×10^{-3} μc/ml for alpha activity(4). The values mentioned above are summarized in Table 1 along with values given for the more important radioisotopes in use(3). Morgan and Straub(5) have presented equations for calculating permissible levels of activity as a function of the time after a nuclear detonation.

The per cent removal that must be provided by a particular treatment process is a function of the initial concentration of the radioactive element and the MPC value for that radioisotope as given in NBS Handbook 52. If for example, the initial concentration of strontium-89 in water amounts to 1 μc/ml (or 3×10^{-5} ppm), the removal required to reduce the activity to the MPC value of 7×10^{-5} μc/ml (or 2×10^{-9} ppm) is 99.993 per cent, a very high removal. Or, if the efficiency of a particular process and the MPC value are known, the maximum concentration of that component in the water for public consumption can be defined. Again using the example cited above, if the process had a removal efficiency of 90 per cent, the maximum concentration of strontium-89 in the untreated water can not be more than 7×10^{-4} μc/ml (or 2×10^{-8} ppm).

Table 1

MAXIMUM PERMISSIBLE CONCENTRATION VALUES IN WATER

ISOTOPE	PERMISSIBLE CONCENTRATION		REMARKS
	μc/ml	mg/l (ppm)*	
Mixed F. P.	10^{-7}		For α, β, and γ - long time use (70 years).
β - γ	9×10^{-2}		For emergency use - 10 days after nuclear explosion.
α	5×10^{-3}		For emergency use - 10 days after nuclear explosion.
Sr^{90}-Y^{90}	8×10^{-7}	4×10^{-9}	For long time use. 0.3 rem/week.
I^{131}	3×10^{-5}	2×10^{-10}	For long time use. 0.3 rem/week.
Sr^{89}	7×10^{-5}	2×10^{-9}	For long time use. 0.3 rem/week.
Cs^{137}-Ba^{137}	1.5×10^{-3}	2×10^{-5}	For long time use. 0.3 rem/week.
Ba^{140}-La^{140}	2×10^{-3}	3×10^{-8}	For long time use. 0.3 rem/week.
Zr^{95}-Nb^{95}	4×10^{-3}	2×10^{-7}	For long time use. 0.3 rem/week.
Ce^{144}-Pr^{144}	4×10^{-2}	1×10^{-5}	For long time use. 0.3 rem/week.
Ru^{106}-Rh^{106}	1×10^{-1}	3×10^{-5}	For long time use. 0.3 rem/week.
P^{32}	2×10^{-4}	7×10^{-10}	For long time use. 0.3 rem/week.
Co^{60}	2×10^{-2}	2×10^{-5}	For long time use. 0.3 rem/week.

* Based on the activity of the parent.

EFFICIENCY OF WATER TREATMENT PROCESSES FOR THE REMOVAL OF RADIOACTIVE CONTAMINANTS

Before speaking specifically of water decontamination and citing laboratory results, attention is called to several important characteristics of radioactive materials. First, the rate of disintegration of radioisotopes cannot be modified by any physical, chemical, or biological processes. In other words, in the treatment processes employed the radioactive materials are transferred from the liquid to the solid phase (chemical precipitates, sludges, or soils). The radioactive materials associated with these solids must still be disposed of in a safe manner. The most common practice is to store these concentrated radioactive solids in tanks or to bury the material in approved areas where there is no likelihood of ground water contamination. Second, chemically, radioactive isotopes react similarly to their stable counterparts. Thus, if a process removes the stable element, as in the case of the precipitation of calcium in softening, radiocalcium will be carried down in the same manner. This statement may be expanded further to include substances such as strontium and barium which are chemically related to calcium, i. e., a process designed for the removal of calcium also will remove some of the strontium and barium. Third, the amounts of the radioelement presently encountered are exceedingly small. To illustrate this a comparison is presented in Table 2 of the permissible levels of specific elements in the Drinking Waters Standards with concentrations indicated for radioisotopes of these elements. It will be seen that the permissible levels from the standpoint of radioactivity are many orders of magnitude below the

Table 2

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QUANTITATIVE COMPARISON OF CHEMICAL CHARACTERISTICS U.S. PUBLIC HEALTH SERVICE DRINKING WATER STANDARDS AND MAXIMUM PERMISSIBLE CONCENTRATION OF RADIOISOTOPES IN WATER

CHEMICAL CONSTITUENT	DRINKING WATER STANDARDS		MAXIMUM PERMISSIBLE CONCENTRATION		RADIOACTIVE FORM
	ppm (mg/l)	μg/cc	μg/cc	μc/cc	
GROUNDS FOR REJECTION (DWS)					
Lead (Pb)	0.1	1×10^{-1}			
Fluoride (F)	1.5	1.5×10^{0}			
Arsenic (As)	0.05	5×10^{-2}			
Selenium (Se)	0.05	5×10^{-2}			
Chromium (Cr^{+6})	0.05	5×10^{-2}	5.2×10^{-4}	0.5	Cr^{51}
RECOMMENDED VALUES (DWS)					
Copper (Cu)	3.0	3×10^{0}	2.1×10^{-5}	8×10^{-3}	Cu^{64}
Iron and Manganese (Fe + Mn)	0.3	3×10^{-1}	1.8×10^{-6} 2.1×10^{-9}	4×10^{-3} 1×10^{-4}	Fe^{55} Fe^{59}
Magnesium (Mg)	125	1.25×10^{2}	9.3×10^{-9}	0.2	Mn^{56}
Zinc (Zn)	15	1.5×10^{1}			
Chloride (Cl)	250	2.5×10^{2}	8.9×10^{-2}	2×10^{-3}	Cl^{36}
Sulfate (SO_4)	250	0.83×10^{2} (as sulfur)	1.2×10^{-7}	5×10^{-3}	S^{35}
Phenolic compounds* (C_6H_5OH)	0.001	1.5×10^{-4}*	6.7×10^{-4}	3×10^{-3}	C^{14}

*Assuming that only one of the carbon atoms is radioactive C^{14}.

concentrations permitted on the basis of the Drinking Water Standards. Fourth, being present in such low concentrations usual qualitative and quantitative procedures cannot be used in their identification. However, because these materials are radioactive, this characteristic may be utilized to detect, measure, and identify them. To determine specific radioisotopes, radiochemical procedures must be employed and these become less accurate as the number of radioactive constituents present increases.

Conventional water treatment processes include coagulation, settling, filtration, and disinfection. In addition, auxiliary treatment such as aeration, softening, iron and manganese removal may be provided. In the course of the experimental studies carried out at the Oak Ridge National Laboratory by the Laboratory, the Public Health Service, the Engineer Research and Development Laboratory, Fort Belvoir, and the Tennessee Valley Authority, non-conventional processes were investigated also and included clay adsorption, phosphate coagulation, adsorption on metal powders, and electrodialysis with permselective ion exchange membranes.

Coagulation. Coagulation involves the formation of chemical flocs that adsorb, entrap, or otherwise bring together suspended matter, particularly suspended matter that is colloidal. The addition of coagulating chemicals will result also in the precipitation of some of the soluble constituents in the waste as metal hydroxides, generally hydroxides of aluminum or iron, since alum and iron salts are the most common coagulants.

Coagulation has been studied rather extensively at the Oak Ridge

National Laboratory, where standardized jar test procedures were used and aluminum sulfate, ferrous sulfate, and ferric chloride were evaluated. To conserve time and effort statistical procedures were employed in planning and carrying out the experiment, since a re-examination and recalculation of an earlier 3^4 factorial experiment, in which the simultaneous effects of turbidity, coagulant dose, pH, and activity were determined, indicated that a 1/3 replication of the 3^4 experiment would provide equally reliable interpretation of the factor interactions. Through the use of this technique it was possible to reduce the number of jar tests from 243 to 54 for each series, or to 2/9 of the original effort. In each series, which included one of the 17 individual radioisotopes and one fission products mixture, experiments were carried out on the simultaneous effect of three levels of activity, three coagulants, three dosages of coagulant, and the day to day variations of the tap water substrate or analyst variation over the three day period of test. The results of these studies are summarized in Table 3, and show that with the exception of most cations of valence 3, 4, or 5, including the rare earths group, coagulation was not very effective for the removal of radioactive materials from water(6)(7). A statistical analysis of the reported data indicated the interrelationships shown in Table 4. The values reported give the probability that the observed interactions were due to chance. A probability value of <0.001 was taken as the level of significance. In many cases it is customary to assume a probability value of 0.05 as significant. It will be seen that the significance of the four factors evaluated varied with the individual radioisotopes studied.

Table 3

REMOVAL OF RADIOACTIVITY - CHEMICAL COAGULATION

(Ferrous sulfate, ferric chloride, and aluminum sulfate at 1, 2, and 6 gpg)[1]

Isotope	No. of Tests	Activity Range c/m/ml	pH Range		Per Cent Removal	
			Initial	Final	Average	Range
Cs^{137}-Ba^{137}(Cl)	27	19-17,390	7.5-7.8	6.5-7.8	0.5	0-37
$Sr^{89}(Cl_2)$	54	212-25,950	7.7-7.9	7.5-8.0	3	0-15
Ba^{140}-$La^{140}(Cl_2)$	81	42-17,150	7.4-7.6	7.2-8.2	59	1-84
	54	42- 5,520	7.4-7.6	6.6-7.9	44	19-59
	54	44- 8,070	7.4-7.6	7.2-7.8	52	27-72
$Sc^{46}(Cl_3)$	27	163-24,300	7.2-7.8	6.4-8.0	89	62-98
	27	326-20,060	7.6	7.0-7.8	92	62-99+
	27	202-26,100	7.5-7.9	6.9-8.2	91	69-99+
$Y^{91}(Cl_3)$	54[2]	188- 4,166	7.5-7.6	6.0-7.6	90	40-99
	81[2]	367-20,860	8.1	7.8-10.2	92	56-99+
	81[2]	202-21,450	7.8-8.0	7.6-8.4	86	48-99
	81[3]	406-21,500	7.8	7.5-9.1	75	1-98
	81[3]	222-21,500	7.1-8.4	7.4-9.5	73	33-98
Zr^{95}-Nb^{95}(oxylate complex)	54	186-27,200	7.4-7.6	6.4-7.9	80	2-99
P^{32} as PO_4	27	228-17,625	7.6	7.4-7.8	96	68-99+
$Cr^{51}(Cl_3)$	54	5- 758	6.7-7.3	5.3-7.8	6	0-60
$Mo^{90}(O_3)$	54	84-10,085	7.6	7.0-8.2	10	0-60
W^{185}(tungstate)	54	233-28,290	7.5-7.9	6.6-8.4	46	1-96
	81	198-22,490	8.0-8.1	7.5-8.4	36	4-91
Re^{186}(metal)	54	95-15,230	7.6	7.2-8.0	6	0-29
I^{131}(iodide)	27	239-25,060	7.6	7.2-7.8	20	0-44
$Ru^{103}(Cl_3)$	27	196-24,960	7.3-7.6	6.9-8.2	77	43-96
Pr^{142}(as Pr_2O_3)	27	102-12,050	7.3	7.4-8.0	96	83-99+
Ce^{144}-$Pr^{144}(Cl_3)$	27	244-30,510	7.4-7.6	7.2-8.4	91	28-99+
$Pm^{147}(Cl_3)$	27	136-19,040	7.4-7.6	7.0-8.2	87	4-99+
Sm^{153}(as Sm_2O_3)	54	148-20,870	7.4-7.6	5.8-8.0	91	44-99+
F. P. Mixture	27	216-32,320	7.4-7.6	7.4-8.2	73	46-86

1 1 gpg means 1 gpg of either $FeSO_4$, $FeCl_3$ or alum + 0.4 gpg activated sodium silicate + 1 gpg of $Ca(OH)_2$ or Na_2CO_3. 2 gpg and 6 gpg are multiples of these amounts.

2 $Ca(OH)_2$ was used as alkaline agent.

3 Na_2CO_3 was used as alkaline agent.

Table 4

RESULTS OF ½ REPLICATION OF 3^4 FACTORIAL EXPERIMENT – COAGULATION STUDIES

Significance of Day to Day Variations (A), Activity Level (B), Coagulant (C), and Dose (D) as Well as Their Interrelationships*

Isotope	Level of Significance P =										Remarks
	A	B	C	D	AB	AC	AD	BC	BD	CD	
Co^{137} (Cl)	>0.20	>0.20	>0.20	>0.20	>0.20	>0.20	>0.20	>0.20	>0.20	>0.20	Not significant
$Sr^{89}(Cl)_2$	>0.20	0.01-0.05	0.20	<0.001	0.001-0.01	>0.20	>0.20	0.05-0.10	>0.20	>0.20	D significant
$Ba^{140}(Cl)_2$	0.001-0.01	0.05	<0.001	<0.001	0.10-0.20	>0.20	0.01-0.05	<0.001	0.05-0.10	<0.001	C, D, BC, and CD significant
$Y^{91}(Cl)_3$	<0.001	<0.001	<0.001	<0.001	0.10-0.20	<0.001	>0.20	<0.001	>0.20	<0.001	A, B, C, D, AC, BC, and CD significant
Zr^{95} (oxy)	<0.001	<0.001	<0.001	<0.001	0.01-0.05	0.05-0.10	>0.20	0.001-0.01	0.01-0.05	0.001	A, B, C, and D significant
Cr^{51} (metal)**	>0.20	<0.001	0.05-0.10	0.10-0.20	0.10-0.20	>0.20	>0.20	>0.20	0.10-0.20	>0.20	B significant
Mo^{99} (metal)	0.001-0.01	<0.001	<0.001	<0.001	0.001-0.01	0.01-0.05	0.001-0.01	0.10	<0.001	<0.001	B, C, D, BD, and CD significant
$K_2W^{185}O_4$	<0.001	<0.001	<0.001	<0.001	>0.20	<0.001	0.001-0.01	<0.001	<0.001	<0.001	A, B, C, D, AC, BC, BD, and CD significant
Re^{186} (metal)	<0.001	<0.001	>0.20	0.01-0.05	>0.20	>0.20	>0.20	>0.20	>0.20	>0.20	A and B significant
$Ru^{103}(Cl)_2$	0.05-0.10	<0.001	<0.001	<0.001	>0.20	>0.20	>0.20	>0.20	>0.20	0.01-0.05	B, C, and D significant
$P^{32}O_4^{=}$	Not sig.	Sig.	Slightly sig.	Slightly sig.	Not sig.	Not sig.	Not sig.	Not sig.	Not sig.	Not sig.	B significant
NaI^{131}	Sig.	Not sig.	Sig.	Not sig.	Not sig.	Sig.	Not sig.	Not sig.	Not sig.	Not sig.	A, C, and AC significant
P_r^{142} (metal)	Not sig.	Sig.	Not sig.	Sig.	Not sig.	Not sig.	Not sig.	Not sig.	Not sig.	Not sig.	B and D significant
$Sn_2^{113}O_3$	0.10-0.20	0.001-0.01	>0.20	0.01-0.05	>0.20	>0.20	>0.20	>0.20	0.20	>0.20	B most significant

*A = day by day variation (over three day period), B = activity level – 200, 2,000, and 20,000 c/m/ml (uncorrected), C = coagulants ($FeSO_4$, $FeCl_3$ or alum), and D = dose (1, 2, or 3 grains per gallon which includes primary coagulant, 40 per cent sodium silicate, and lime).

**Cr^{51} activity levels were 50, 100, and 200 c/m/ml (uncorrected).

Phosphorus-32 has been removed from waters contaminated with this radioisotope[7][8], but considerable care had to be given to the coagulation step[8]. Removals in excess of 98 per cent have been reported. Iodine-131, which has found most extensive use in medical therapy and research, being in the form of an anion, cannot be removed by coagulation with alum or iron. However, the addition of small amounts of activated carbon[7], copper sulfate[7], or silver nitrate[7][9] has increased removals of this anion from water to about 96 per cent. Some of these results are indicated in Table 5.

Coagulation will be helpful also in the removal of radioactive material attached to or adsorbed on the natural turbidity found in many surface waters. Since coagulation will remove much of the turbidity, the activity attached to it will also be removed. The effect of added turbidity on the removal of certain radioisotopes is illustrated by the data in Table 6.

As many would suspect, pH exerts an appreciable effect on removal. Table 7 shows the effect of pH on the removal of mixed fission products activity and Figure 1 the effect of pH on the removal of ruthenium. Increased chemical dosages do not necessarily result in increased removals of activity. As shown by the data in Table 8 a large increase in the coagulant dose did not result in a proportionate increase in removal of radioactive materials.

Sand Filtration. Except for removal by simple straining or sorption by biological life contained in the "Schmutzdecke", sand filters have not been effective for the removal of radioactive materials[10][11][12]. Their major function is to remove the radioactivity previously incorporated in floc

Table 5

REMOVAL OF I^{131} BY AUXILIARY PROCESSES

Dosage				Final Characteristics			Removals - Per Cent		Source of Data
Turbidity	Alum	NaOH	Auxiliary Chemical	pH	Alkalinity	Turbidity	Coagulation and Settling	Coagulation Settling and Centrifuging	
ppm					ppm				
103	25.7	38.9	5-15[a]	5.8-7.1	17-68	0-15	64-72	74-78	(7)
103	25.7	38.9	3.5[b]	7.1	65	0	38	56	(7)
17.3-274	25.7	38.9	0.12-3.8[c]	5.8-7.5	19-74	1-40	42-74	48-76	(7)
	123.5		0.013-0.13[c]	6.0-6.9			55-90		(9)
	247		0.13-1.1[c]	6.3			90-92		(9)
	100.64[d]		0.13	6.3			95.5		(9)

[a] Auxiliary chemical was activated carbon.

[b] Auxiliary chemical was copper sulfate.

[c] Auxiliary chemical was silver nitrate.

[d] 20 ppm as Fe or 100.64 ppm as $Fe_2(SO_4)_3 \cdot 9\ H_2O$.

Table 6

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COAGULATION AND SETTLING RESULTS - JAR TEST STUDIES

ISOTOPE	CLAY ADDED (ppm)[a]	COAGULANT ADDED (gpg)[b]	FINAL pH	PER CENT REMOVAL
Cs^{137}-Ba^{137}	0	1	-	0- 6
	100	1	-	35-65
Sr^{89}	0	1.5*	6.7- 7.8	0- 6
	100	0.5 - 6	6.7-10.7	0-51
Cd^{115}	0	1	-	40-60
	100	1 - 5	-	60-95
Ba^{140}-La^{140}	100	1 - 6	7.5- 8.2	28-84
Sc^{46}	100	1 - 6	6.5- 8.2	66-98
Y^{91}	0	1.5*	6.8- 7.1	83-93
	100	1 - 6	7.0-10.2	34-99
Zr^{95}-Nb^{95}	0	1 - 5	-	70-98
	100	1	-	95-99
P^{32}	100	0.5 - 1.5*	6.8- 8.8	97-99
Cr^{51}	100	1 - 6	7.6- 8.8	73-98
W^{185}	100	1 - 6	7.5- 8.4	5-91
I^{131}	100	0.5 - 2*	6.9- 9.0	0-10
Ce^{144}	0	1 - 1.5*	7.2- 7.8	81-94
	100	0.5 - 2.5*	7.0- 7.8	85-96
FPM-1	100	1 - 3*	7.2- 8.8	61-84
FPM-2	0	1 - 5	4.3-10.2	9-71
	100	1 - 5	4.3-10.2	12-73
FPM-3	0	10	9.9-10.0	46

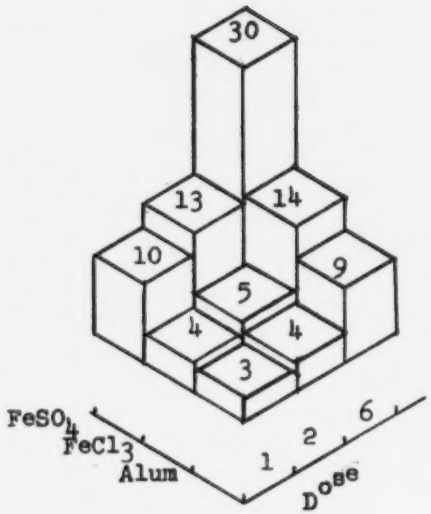
[a] Local clay was added,
[b] Coagulant includes alum, ferrous sulfate, or ferric chloride, lime, soda ash, or sodium hydroxide, and sodium silicate.
* No sodium silicate added. Where added sodium silicate equals to 40% primary coagulant dose.
FPM-1 - Iodine dissolver solution.
FPM-2 - Synthetic mixture containing fission products in the same concentrations assumed to be present 30 days after an under water bomb blast.
FPM-3 - Three year old fission product mixture.

Table 7

EFFECT OF INCREASED pH ON THE REMOVAL OF MIXED FISSION PRODUCTS BY FILTRATION

pH	Per Cent Removed by Filtration
2.5	14.6
6.2	63.1
8.6	80.7
10.5	85.1
12.2	88.6

pH 2.5-4.4



pH 7.5-8.4

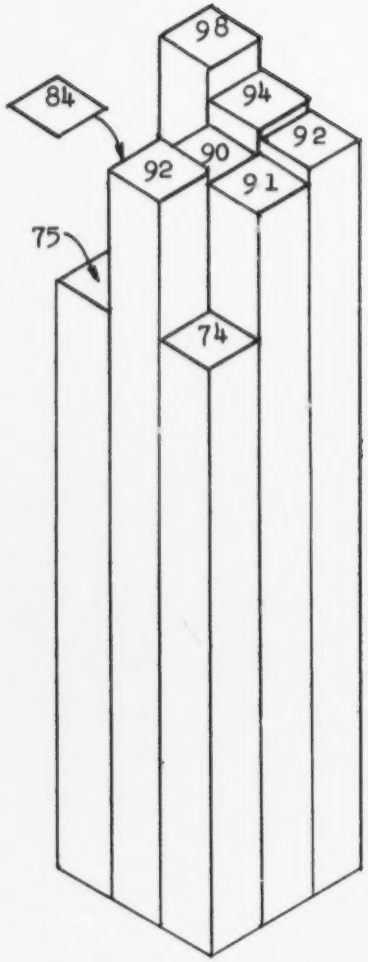


Figure 1. Effect of pH on Removal of Ru^{103}

Table 8

EFFECT OF INCREASING DOSES OF IRON ON THE

REMOVAL OF MIXED FISSION PRODUCTS

$FeCl_3 \cdot 6\ H_2O$ ppm	Per Cent Removal
10	77.9
20	80.3
30	81.5
40	82.4
70	83.7
120	85.3
200	87.5
270	88.6

particles. As shown in Table 9 removals vary depending on the nature of the materials. For example, scandium, yttrium, and zirconium-niobium which were probably present in the colloidal state were removed by mechanical straining, whereas other materials such as strontium, cesium, and possibly tungsten, which were present in true solution were not effected by passage through the sand filter.

Softening. Softening with lime and soda ash has been studied and under proper conditions effective removals of strontium, barium, cadmium, yttrium, scandium, and zirconium-niobium have been obtained[12][13][14][15]. As shown in Table 10, removals of 95 to 99 per cent or better are possible. For most satisfactory removal of strontium, Hoyt[13] found that excesses of both lime and soda ash were required. His results are summarized in Table 11. The strontium present was reported to be removed by coprecipitation with the calcium carbonate by the mechanism of mixed crystal formation[13][15]. Data on the removal of stable strontium by municipal water treatment processes including softening have been reported[16] and are reproduced in Table 12.

As has been indicated above, lime-soda softening is fairly effective in removing certain radioelements if relatively high chemical dosages are employed. From the standpoint of emergency water treatment, this process is probably of little concern in that water works serving less than 15 per cent of this country's population are presently equipped to soften water. Also, exceedingly high dosages would be required to effect substantial reduction of mixed fission products. The softening process could be adapted

Table 9

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LABORATORY SAND FILTRATION RESULTS

ISOTOPE	INITIAL ACTIVITY c/m/ml	VOLUME PASSED ml	pH OF EFFLUENT	PER CENT REMOVAL	
				RANGE	AVERAGE
Cs^{137}-Ba^{137}	800	500	8.3	10-70	50
Sr^{89}	2,700	750	8.3	1-13	4
Cd^{115}	1,200	500	9.1	60-99	95
Ba^{140}-La^{140}	1,300	750	7.6	39-99	74
Sc^{46}	1,500	750	8.3	94-99	96
Y^{91}	5,700	750	7.0	84-89	87
Zr^{95}-Nb^{95}	3,400	500	7.8	91-96	93
W^{185}	2,200	750	7.1	3-18	8

Table 11

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APPROXIMATE MINIMUM COMBINED DOSAGE* OF LIME AND SODA ASH TO GIVE STATED REMOVAL

ISOTOPE	CHEMICAL DOSAGE IN gpg FOR STATED PER CENT REMOVAL									
	50		75		90		95		99	
	LIME	SODA	LIME	SODA	LIME	SODA	LIME	SODA	LIME	SODA
$Ba^{140} - La^{140}$	2	2	4	2	6	4				
Sr^{89}	4	3	5	5	7	9	20	20		
Cd^{115}	2	2	3	3	4	4	4	4	6	4
Sc^{46}	3	3	3	3	5	5				
Y^{91}	2	2	4	4	6	6	12	6		
$Zr^{95} - Nb^{95}$	2	0	5	0	12	0	17	0	22	0
Cs^{137}	48	48								
W^{185}	48	48								

* MINIMUM COMBINED DOSAGE IS DEFINED SUCH THAT, OF THOSE VARIABLE DOSAGES STUDIED, THE gpg LIME PLUS THE gpg SODA ASH IS A MINIMUM.

Table 11

RESULTS OF LIME-SODA ASH TREATMENT FOR REMOVAL OF STRONTIUM

Treatment	Per Cent Removal of Activity
Stoichiometric amounts	75.0
20 ppm excess lime-soda ash	77.0
50 ppm excess lime-soda ash	80.1
100 ppm excess lime-soda ash	85.3
150 ppm excess lime-soda ash	97.3
200 ppm excess lime-soda ash	99.4
300 ppm excess lime-soda ash	99.7

Table 12

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STRONTIUM REMOVAL BY CHEMICAL TREATMENT

TYPE OF TREATMENT	SR REMOVAL–PER CENT		NO. OF CITIES
	AVERAGE	RANGE	
ALUM OR FERROUS SULFATE	12	10–31	7
ALUM OR FERROUS SULFATE, PLUS LIME	37	10–75	11
ALUM OR FERROUS SULFATE, PLUS LIME AND SODA ASH	54	10–85	3
ALUM OR FERROUS SULFATE, PLUS LIME AND PHOSPHATE	42	10–70	5
SOFTENING ONLY (PHOSPHATE, ION EXCHANGER)	73	69–76	2
NONE (EXCEPT CHLORINE, FLUORIDE, CARBON, OR AMMONIA)	10		8

to treatment of a waste containing radiostrontium. Other water treatment processes, with the exception of ion exchange, are relatively ineffective in removing this particular element, whereas softening, under optium conditions can effect removals in excess of 99 per cent.

Ion Exchange. Another method of softening used in muncipal water treatment practice is ion exchange. Removal of radioactive materials by this process is technically possible[12][14][17][18], and for most effective removals mixed bed ion exchange resins in series are recommended. Some results with various types of exchange materials are shown in Table 13, and are compared with removals obtained with greensands[12], which are used in municipal softening installations. In these studies the cation exchange materials evaluated included IR-120 (H^+ and Na^+ cycles) and a natural greensand (Na^+ cycle); the anion exchangers included Dowex -1 (OH^- and Cl^- cycles); and the mixed beds included an IR-120 (H^+) - Dowex - 1 (OH^-) bed and an IR-120 (Na^+) - Dowex - 1 (Cl^-) bed. In general, the particular regeneration cycle, i.e., whether H^+ or Na^+ regenerated cation exchange resin of OH^- or Cl^- regenerated anion exchange resin, did not appreciably influence the results. In water treatment practice, the effluent pH may be of significance. The range in effluent pH for the various materials studied was as follows: IR-120 (H^+) 2.8-3.6; IR-120 (Na^+) 3.4-4.3, Dowex - 1 (OH^-) 10.7-11.2; Dowex - 1 (Cl^-) 7.2-8.7; greensand 7.1-8.0. The removal efficiencies with greensand do not appear to be significantly different from the sodium-cycle cation exchange resin results; however, the total capacities of the resins undoubtedly differ greatly.

Table 13

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PERCENTAGE REMOVAL RESULTS OF INDIVIDUAL ISOTOPES WITH ION EXCHANGE MATERIALS

ISOTOPE	ION EXCHANGE MATERIAL			
	CATION EXCHANGE	ANION EXCHANGE	MIXED BED	GREENSAND
W^{185}	12 -16	97.2-99.2	98.9	9
Y^{91}	86 -93.1	94.2-98.5	97.6 -98.7	75
Sc^{46}	95.7-97.2	98.8-99.0	98.5 -98.7	96.4
Sr^{89}	99.1-99.8	5 - 7	99.95-99.97	99.8
Ba^{140} - La^{140}	98.3-99.0	36 -42	99.5 -99.6	96.3
Cs^{137}	99.8	9	99.8	--
Cd^{115}	98.5	0	99.2	--
Zr^{95} - Nb^{95}	58 -75	96.4-99.9	90.9 -99.4	--

It will be noted that strontium, cesium, cadmium, and, to a lesser extent, barium-lanthanum are present in solution as cations, whereas tungsten is present as an anion. Yttrium, scandium, and zirconium-niobium are apparently in colloidal form and, as such, are removed to some extent on both cation and anion resins.

A more extended study of ion exchange was made by Friend[17], who investigated this procedure for the removal of I^{131}, Sr^{89}, and mixtures of the two radioisotopes. He reported a decontamination factor (influent activity divided by effluent activity) in excess of 10^2 (>99 per cent removal) for carrier-free I^{131} through the use of IRA-400 resin (OH^- cycle) or a combination of IR-120 (Na^+ or H^+ cycles) and IRA-400 (OH^-cycle); a decontamination factor in excess of 10^3 (>99.9 per cent removal) for carrier-free Sr^{89} through the use of IR-120 (H^+ cycle) or a combination of IR-120 (H^+ or Na^+ cycles) and IRA-400 (OH^- cycle); and a decontamination factor in excess of 10^3 (>99.9 per cent removal) for the carrier-free mixture of Sr^{89} and I^{131} through the use of IR-120 (H^+ or Na^+ cycles) and IRA-400 (OH^- cycle).

Further studies on the use of mixed cation and anion exchange resin (Amberlite MB-3) slurries for the removal of a variety of radioisotopes from water solution have been made by Lacy and are summarized in Table 14.

Although the ion exchange process offers one of the most promising methods for removing radio-contaminants, the cost may preclude widespread usage unless suitable regeneration techniques are developed.

Table 14

REMOVAL OF RADIOACTIVE CONTAMINANTS FROM WATER

BY COMBINED ANION-CATION RESIN

RESIN-JAR TEST STUDIES (STIRRING TIME 90 MINUTES, SAMPLES FILTERED)

Contaminant	Initial pH	Initial Activity c/m/ml	Per Cent Removal Mixed Ion Exchange Resin - ppm			
			450	900	1800	2700
P^{32}	8.2	5,560	47.4	74.5	96.2	99.8
Cd^{115}	8.0	7,880	37.9	45.6	91.1	99.99
Cs^{137}-Ba^{137}	8.2	8,200	15.1	14.6	69.1	99.99
Zr^{95}-Nb^{95}	8.1	6,700	98.3	98.4	99.2	99.4
I^{131}	7.5	3,200	84.5	93.5	95.6	98.1
$Ce^{141,144}$-Pr^{144}	7.9	4,150	98.7	99.2	99.8	99.98
Ba^{140}-La^{140}	7.6	3,490	85.1	94.5	98.8	99.9
FPM-4	8.3	13,600	82.7	90.5	97.3	99.2
FPM-5	2.7	3,400	38.4	----	----	----

FPM-4 - Iodine dissolver solution aged 30 days.
FPM-5 - Mixed fission product waste containing mainly Cs^{137}-Ba^{137} and Ru^{106}-Rh^{106}.

Modified Processes

Several other methods have been studied for the removal of radioactive constituents from water. The most promising of these, phosphate coagulation[19][20], has effected removals up to 99.9 per cent as shown by the data included in Table 15. Relatively high removals of strontium are also possible with this method, but these can be secured only under carefully controlled conditions of pH and the calcium hydroxide to trisodium phosphate ratio as illustrated in Figure 2.

Studies utilizing an electrodialytic cell with permselective ion exchange membranes[21] indicated the necessity of removing all colloidal radioelements prior to concentration of the soluble radioisotopes in the cell. Passage of various radiocolloids through a millipore filter has shown that the larger colloids such as Zr^{95}-Nb^{95} were removed completely, whereas others such as Ru^{103} and Ce^{144}-Pr^{144} were removed only in part. Therefore, if the dialytic cell is to be effective for the removal of the soluble radioactive materials, the radiocolloids should be removed beforehand.

Other methods suggested for the removal of radioactive materials include the use of metal powders and added clay materials. Laboratory jar test studies were carried out by Lacy[22] for the removal of specific radioactive contaminants (as shown in Table 16) by various metal dusts. In general, the highest removals were obtained with the iron dust and exceeded 90 per cent, except in the cases of the fission products mixture, iodine, and cesium.

Table 15

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REMOVAL OF RADIOISOTOPES BY PHOSPHATE COAGULATION

ISOTOPE	COAGULANT	COAGULANT DOSE mg/liter	REMOVAL PER CENT
Ce^{144}	KH_2PO_4	200	99.8
		120	99.9
	Na_3PO_4	100	81.3
Sr^{89}	KH_2PO_4	240	97.8
	Na_3PO_4	100	99.9
Y^{91}	KH_2PO_4	100	66.1
Sb^{124}	KH_2PO_4	120	67.4
	Na_3PO_4	50	99.6
Zn^{65}	KH_2PO_4	200	10.7
W^{185}	KH_2PO_4	100	99.5
Zr^{95}	KH_2PO_4	100	99.2
Nb^{95}	KH_2PO_4		

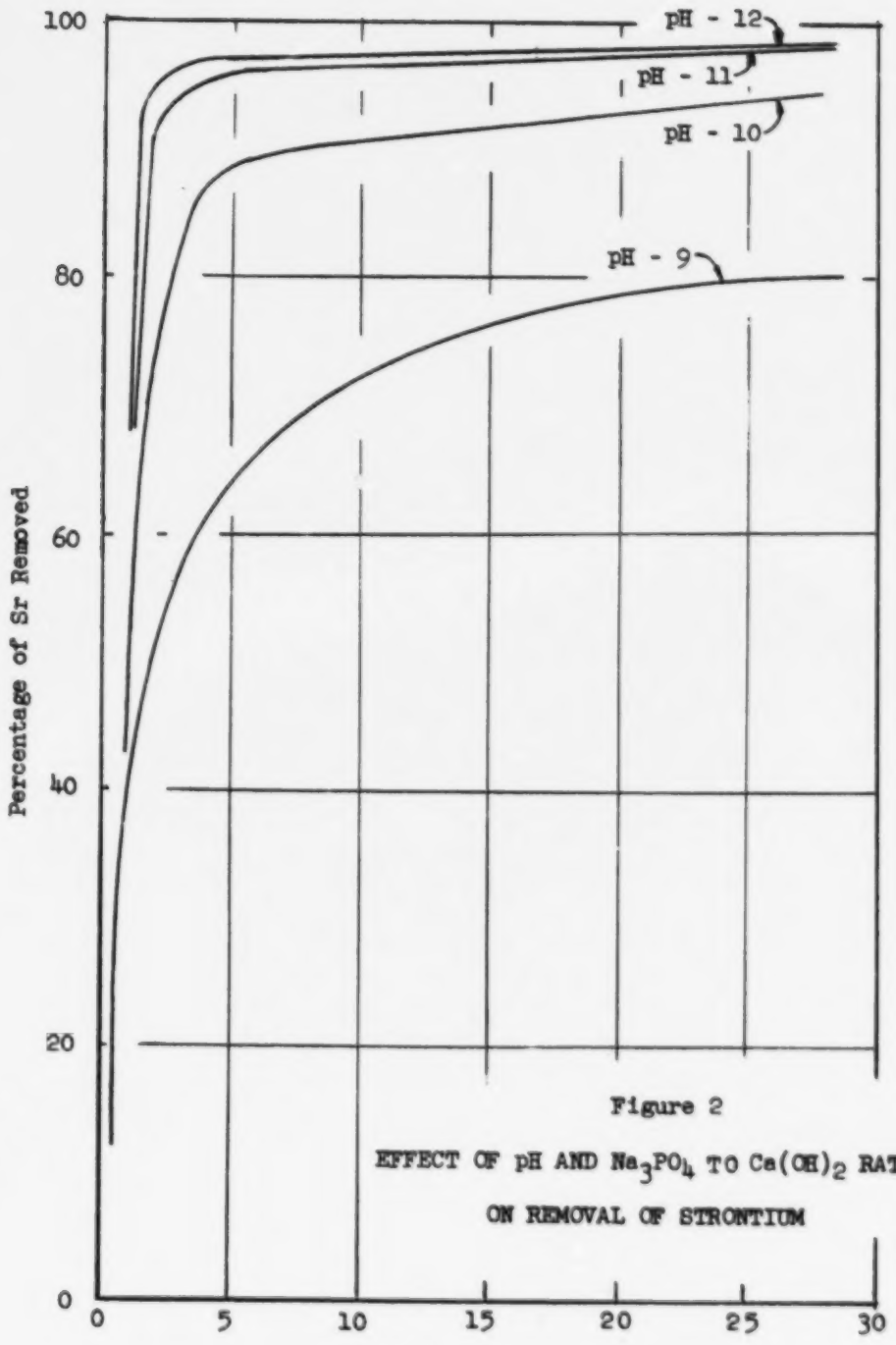

Figure 2

EFFECT OF pH AND Na_3PO_4 TO $Ca(OH)_2$ RATIO ON REMOVAL OF STRONTIUM

Table 16

Unclassified
Dwg. 21776

REMOVAL OF RADIOACTIVE CONTAMINANTS BY METALLIC DUSTS
(1000 ppm Metal Dust, 90 Minute Contact, Filtered Sample Counted)

CONTAMINANT	STOCK SOLUTION pH	INITIAL ACTIVITY c/m/ml	PER CENT REMOVAL			
			Fe[a]	Zn	Cu	Al
Ru^{106}-Rh^{106}	8.0-8.2	1,975 - 3,950	99.6	98.4	93.7	92.8
Y^{91}	7.9-8.0	1,610 - 3,230	98.0	98.0	---	---
Zr^{95}-Nb^{95}	7.2-8.3	1,210 - 4,470	99.1	97.9	99.1	99.8
P^{32}	7.8-8.4	128 - 12,000	99.8	98.1	---	84.2
I^{131}	6.5-8.2	4,288 - 6,689	37.2	45.7	42.2	23.2
$Ce^{141,144}$-Pr^{144}	7.9-8.1	3,915 - 4,491	99.9	99.9	99.5	99.8
Ba^{140}-La^{140}	7.9-8.2	4,644 - 10,205	94.8	74.5	65.7	73.8
Co^{60}	8.0-8.2	1,571 - 4,792	92.2[b,c]	---	47.8	30.2[b]
FPM-6	7.2-8.0	4,730 - 10,375	85.8	76.6	92.1	89.4
FPM-5	2.0-7.2	2,890 - 4,220	55.5	39.6	49.3	8.2
Cs^{137}-Ba^{137}	8.0	13,085	8.6	---	---	---

[a] Fe samples were centrifuged before counting
[b] 500 ppm metal dust
[c] 60 minute contact
FPM-6 - Iodine dissolver solution aged 20 days
FPM-5 - Mixed fission product waste containing mainly Cs^{137}-Ba^{137} and Ru^{106}-Rh^{106}.

From these results Lacy concluded that the primary mechanism involved in the removal of these radiocontaminants was adsorption. However, other mechanisms such as metallic displacement may be responsible for some of the removals reported.

Considerable work has been reported[7][23][24][25] on the use of clay materials for the removal of specific radioisotopes from water and waste solutions. Some results are indicated in Table 17. Although this method is feasible, large volumes of clay must be handled both initially and as a contaminated mass of material. Furthermore, if combined with chemical coagulation, still larger volumes of radioactively contaminated sludge must be handled for disposal. This method has its greatest potential use where geological and hydrological conditions are such that the ground itself may be used for disposal, providing no health hazard will result from the release of these materials.

Evaporation

Although not a municipal treatment method, evaporation is included in this discussion because it is the most effective process presently available for concentrating the radioactive materials. Under proper conditions it is possible to produce a condensate very low in activity. Thus, high decontamination factors are possible, but the cost is great. However, under emergency conditions evaporation should prove very satisfactory.

Table 17

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EFFECT OF ADDED CLAY* ON REMOVAL OF RADIOISOTOPES

RADIOISOTOPE	PER CENT REMOVAL		
	100 ppm	750 ppm	5000 ppm
Ba^{140} - La^{140}	41	58	85
Cd^{115}	3	30	64
Ce^{144}	70-80	86	--
Cs^{137}	38	87	98.0
Sc^{46}	53	91.7	96.9
Sr^{89}	2-12	14-22	49-52
W^{185}	0	4	49
Y^{91}	22-45	56-70	93.6
Zr^{95} - Nb^{95}	93.5	99.0	98.0

*LOCAL CLAY ADDED TO PRODUCE TURBIDITY.

Removal of Mixed Fission Products

Up to this point the discussion has centered on the removal of specific radioisotopes from waste solutions. In the case of a fission products mixture, or any mixture of radioisotopes, the efficiency of removal is a function of the individual radioisotopes comprising the mixture. Removals to date have been in the order of 70 to 80 per cent with alum or iron coagulation(7). A series of studies on actual radioactive process waste waters at the Oak Ridge National Laboratory disclosed that removals up to 90 per cent could be obtained with excess lime-soda ash softening and phosphate coagulation when coupled with the addition of clay, which was added for the removal of the cesium in the waste(26). Results of laboratory tests with this waste material are summarized in Table 18. Other experimental data with fission products mixtures will be found in Table 19.

REMOVAL OF FALL-OUT MATERIALS

No discussion of water decontamination is complete without some consideration of the fall-out from nuclear weapons tests. While many of the radioactive materials discharged directly into surface or ground waters are in solution, some of these, deposited during fall-out, may be in the form of insoluble oxides, and removal of these may differ from the removal of the soluble ions.

Several studies have been reported recently dealing with the problem of fall-out in particular areas(27)(28)(29)(30)(32). At Albany, New York, it was reported(27) that about 35 per cent of the activity due to fall-

Table 18

LABORATORY STUDIES ON REMOVAL OF RADIOACTIVITY FROM ORNL PROCESS WASTES

Type of Sample	Number of Observations	Treatment	Per Cent Removal	
			Average	Range
Grab*	5	Alum coagulation	28	23-32
Grab*	5	Ferrous sulfate coagulation	32	15-44
Grab*	5	Phosphate coagulation	73	66-82
Grab*	4	Excess lime-soda ash	70	61-78
Grab*	1	Variable clay (100-1,000 ppm)		39-52
Grab	20	Phosphate coagulation	70	43-83
8-hr composite	6	Phosphate coagulation	70	46-78
8-hr composite	6	Excess lime-soda ash	37	19-48
8-hr composite	7	Serial treatment - clay + phosphate coagulation	90	82-93
8-hr composite	6	Serial treatment - clay + excess lime-soda ash softening	84	82-88

* Preliminary tests.

Table 19

UNCLASSIFIED
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REMOVAL OF MIXED FISSION PRODUCTS ISOTOPES

MIXTURE *	TREATMENT PROCESS	PER CENT REMOVAL
FPM - 1	COAGULATION AND SETTLING PLUS CLAY	61 - 84
FPM - 2	COAGULATION AND SETTLING	9 - 71
	COAGULATION AND SETTLING PLUS CLAY	12 - 73
	COAGULATION AND SETTLING PLUS CLAY	46
	COAGULATION AND SETTLING PLUS FILTRATION PLUS CLAY	70 - 73
FPM - 3	COAGULATION AND SETTLING	46
FPM - 4	COAGULATION AND SETTLING	89
	MIXED CATION-ANION EXCHANGE SLURRY	83 - 99
FPM - 5	COAGULATION AND SETTLING	51 - 59
	MIXED CATION-ANION EXCHANGE SLURRY	38
	METALLIC DUST SLURRY	8 - 56
FPM - 6	METALLIC DUST SLURRY	77 - 92

*FPM - 1 IODINE DISSOLVER SOLUTION
FPM - 2 SYNTHETIC MIXTURE
FPM - 3 THREE YEAR OLD FISSION PRODUCTS MIXTURE
FPM - 4 IODINE DISSOLVER SOLUTION AGED 30 DAYS
FPM - 5 MIXED FISSION PRODUCT WASTE CONTAINING Cs^{137}-Ba^{137} AND Ru^{106}-Rh^{106}
FPM - 6 IODINE DISSOLVER SOLUTION AGED 20 DAYS.

out was removed in passing through the Albany water supply system and treatment plant, which included alum coagulation, settling, and filtration, whereas studies reported by Thomas and co-workers[28][29] covering operation of the Lawrence, Massachusetts water treatment plant showed an overall removal of 80 per cent following coagulation, settling, and filtration. Bell et al.[29] compared fall-out removals from the Cambridge and Lawrence, Massachusetts and Rochester, New York water treatment plants with pilot plant studies reported by Straub[7][31] for a simulated bomb mixture one month after the detonation but containing larger amounts of radioactive ruthenium, iodine, and strontium, which are hard to remove by coagulation. Bell reports considerably lower removals of the fall-out activities by water treatment processes than those reported for the simulated mixture. Some data reported by personal letter to the writer[32] following rains on May 17, 1954 in Kyoto City, Japan showed that the water treatment plant, which included settling and filtration (coagulation not being mentioned), removed 34 per cent of the activity contained in the impounded water. Rain water samples containing fall-out materials were collected and analyzed at Cincinnati[30] and indicated that from 50 to 100 per cent of the radioactive materials in the rain water during normal times were soluble, but during the weapons testing program the soluble fraction of radioactive materials was reduced to 30 per cent.

The observations reported above relating to lower removals from surface waters than from simulated or fresh radioactively contaminated solutions are not surprising, since natural factors such as precipitation, adsorption, and assimilation or metabolism by lower animals and plants tend

to remove many of the more easily or readily removable materials leaving behind those materials which are more difficult to remove by conventional water treatment processes.

SUMMARY AND CONCLUSIONS

Unless related to the initial concentration of the radioactive materials present, the per cent removal values cited during the discussion have little significance. It is only when these are considered in terms of the initial concentration and the MPC values that one can be sure that the treated water is safe for human consumption. Generally speaking, water treatment processes will not be effective for reducing the activity in the water to acceptable safe limits, except where the initial levels of activity are very low, certainly several orders of magnitude below the μc/ml range.

Radioactive materials should be retained as close to their source of production as possible, and the practice at present is predominantly tank storage. As pointed out, the transfer of activity from the liquid to the solid phase does not eliminate the problem, since suitable storage or disposal facilities must be provided for the radioactive sludges.

To protect large centers of population, dependent on surface waters which may become contaminated by radioactive materials, plans should be made now for auxiliary sources of water from possible non-contaminated sources. For this purpose existing ground water supplies may be used, other

interconnected water supplies may be piped in, or special provisions may be made to bring water in from the outside. These precautionary measures will apply only in those cases where the external hazard from radioactive materials is sufficiently low that there will be no hazard from this source.

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1/15/55

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